# Neutron-scattering studies of magnetic excitations in La$_{1-x}$Sr$_x$MnO$_3$: A dimensional crossover

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## Abstract

We have found a strong planar ferromagnetic and a weak antiferromagnetic interplane couplings in the spin waves of LaMnO$_3$. Upon doping holes, the anisotropic character initially becomes enhanced. However, it is taken over by the isotropic ferromagnetic spin waves when the Sr concentration $x$ reaches $\sim 0.1$, much lower than the compositional metal–insulator transition range ($x \sim 0.17$).

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Tokura et al. [1] are among the first who pointed out that the colossal negative magnetoresistance (CMR) effects of La$_{1-x}$Sr$_x$MnO$_3$ should be understood in the framework of "strongly correlated electron systems", as apparent in the Mott insulator LaMnO$_3$. Since strong electron correlations affect dynamical spin structures, neutron-scattering experiments will provide key information for the CMR phenomena. We have performed inelastic neutron-scattering experiments on single crystals of La$_{1-x}$Sr$_x$MnO$_3$ ($0 \leqslant x \leqslant 0.3$ [3]), which were prepared by the floating-zone method. In this paper, we refer to a detailed phase diagram determined by Kawano et al. [2] using neutron diffraction. They found a canted antiferromagnetic (CAF) phase between $x = 0$ and 0.15, where the canting angle from the $c$-axis continuously changes from nearly 90° (A-type antiferromagnet) to 0° (ferromagnet), and a pseudo-cubic phase without an obvious Jahn–Teller (JT) distortion (O* orthorhombic phase) in addition to the JT distorted O′ orthorhombic ($x < 0.1$) and the rhombohedral ($x > 0.15$) phases. We have confirmed

that all the samples are consistent with their phase diagram. Measurements were carried out using triple-axis spectrometers; TOPAN at the JRR-3M reactor in Japan Atomic Energy Research Institute and H7 at the high flux beam reactor in Brookhaven National Laboratory.

Fig. 1 shows the spin-wave dispersion of LaMnO$_3$ as well as that of La$_{0.95}$Sr$_{0.05}$MnO$_3$ in the O′ phase at low temperatures. Significant result is a strong, rather two-dimensional (2D), anisotropy in the spin-wave dispersion; a strong planar ferromagnetic (F) coupling along the [$h$ 0 0] and [$h$ $h$ 0] directions, and a weak antiferromagnetic (AF) interplane coupling along the [0 0 $l$] direction. These results are well-described by

$$(\hbar\omega_q)^2 = \{8JS(1 - \cos \pi h \cos \pi k) + 4|J'|S$$
$$+ g\mu_B H_A\}^2 - \{4|J'|S \cos \pi l\}^2, \qquad (1)$$

which is obtained from a simple Heisenberg Hamiltonian with the nearest-neighbor (NN) intraplane F interaction $J$, NN interplane AF interaction $J'$ and the effective anisotropic field $H_A$. The fitting parameters we obtained for $8JS$, $4J'S$ and $g\mu_B H_A$
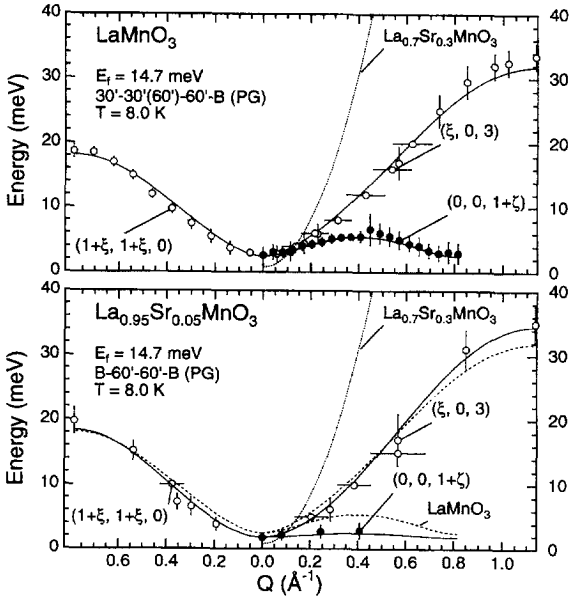
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Fig. 1. The spin-wave dispersion for $x = 0$ and $0.05$. The solid curve corresponds to the fitting with Eq. (1) and the dotted curve is for $La_{0.7}Sr_{0.3}MnO_3$ after Ref. [3].
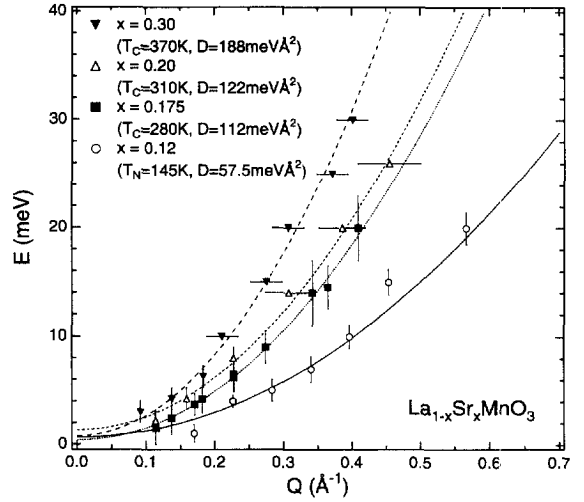


Fig. 2. The spin-wave dispersion for $x = 0.12$, $0.175$, $0.20$ and $0.30$. The spin-stiffness constant $D$ and transition temperature $T_N$ or $T_C$ are indicated.

are 13.3 (2), −4.8 (2), 0.6 (1) meV for $x = 0$, and 16.0 (8), −1.6 (8), 0.8 (4) meV for $x = 0.05$, respectively.

Let us briefly compare with theoretical works. Solovyev et al. [4] explained the stability of the CAF structure of $LaMnO_3$ in the itinerant-electron picture based on the local-spin-density approximation and demonstrated that the $(3x^2 - r^2/3y^2 - r^2)$ type $e_g$ orbital-ordering in the JT distortion plays a crucial role in the magnetism. Their calculation gives $8JS = 4J_{ab}^1/S = 20.1$ meV and $4J'S = (2J_c^1 + 8J_c^2)/S = -10.5$ meV, roughly consistent with experimental values. Ishihara et al. [5], on the other hand, solved an effective Hamiltonian by the mean-field approximation and reached a similar conclusion. They have shown that, with increasing the electron transfer, the F coupling in the $ab$ plane increases while the AF coupling along the $c$ direction decreases. This is consistent with the change of the spin excitations we observed in $LaMnO_3$ and $La_{0.95}Sr_{0.05}MnO_3$.

Fig. 2 shows the spin-wave dispersion for $La_{1-x}Sr_x$ $MnO_3$ with $x \geq 0.12$ at low temperatures. $La_{0.88}Sr_{0.12}$ $MnO_3$ is classified in the CAF insulator and $O^*$ phase

at low temperatures, while the others are in the F metal phase. However, the spin-wave dispersion for $La_{0.88}Sr_{0.12}MnO_3$ shows a very isotropic behavior as seen in the F metal phase; all dispersion curves can be fitted well with $\hbar\omega_q = Dq^2 + E_0$, typical of the F spin-wave dispersion in the low $q$ and low $\omega$ range. Another important finding is that the NN interaction, i.e., $D$ increases with $x$, which coincides with the change in $T_C$ as a function of $x$. We thus conclude that there exists a dimensional crossover from the 2D-like planar ferromagnetic (in the A-type AF ordering) to the 3D isotropic ferromagnetic character at $x \sim 0.1$, where the system still behaves like a semiconductor or insulator. This indicates that the ferromagnetic-metallic feature in the spin dynamics appears in the lower doping level than the compositional metal-insulator transition at $x \sim 0.17$. This crossover agrees with the phase boundary between the $O'$ and $O^*$ structures. In other words, the disappearance of the JT distortion may give rise to this crossover, which is compatible with the theoretical works suggesting that the orbital ordering due to the JT distortion stabilizes the planar F coupling and the A-type AF ordering.

# References

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